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MOLECULAR THEORY OF VISCOELASTICITY FOR HOMOGENEOUS BLOCK AND G--ETC(U)  
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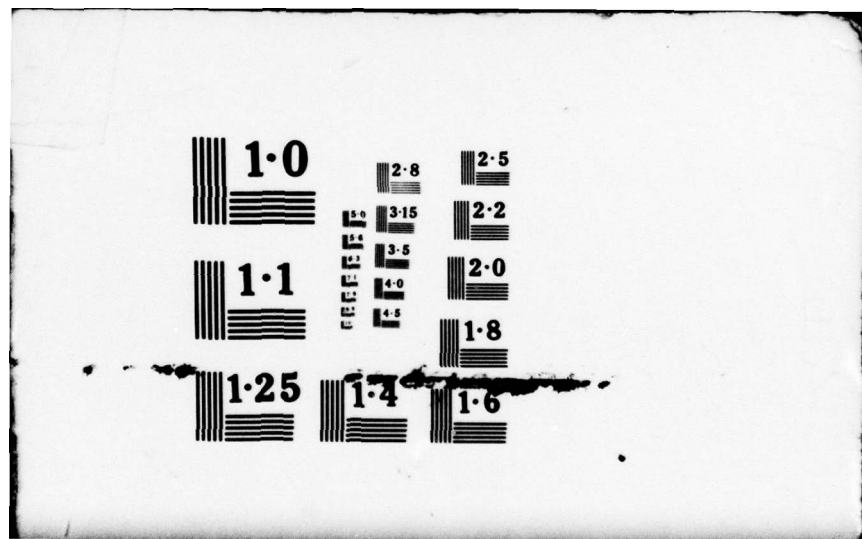
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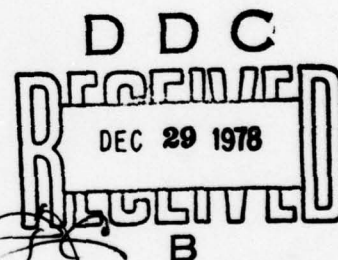
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Molecular Theory of Viscoelasticity for  
Homogeneous Block and Graft Copolymers

by

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Abstract

✓ The Rouse-Bueche-Zimm molecular theory of viscoelasticity has been extended to compute the retardation time spectra of radial block and graft copolymers of various configurations and compositions. The basic assumption of the model is that the effect of the presence of foreign blocks and grafts on the retardation spectrum is due to the difference in the friction coefficients. Mathematically this modification is accomplished by introducing into the equations of motion a frictional coefficient matrix. Retardation spectra can then be obtained by numerically solving for the eigenvalue spectrum on a computer. Results show that the viscoelastic retardation spectra and maximum retardation times of those copolymers vary as a function of the number of side chains, their lengths and the spacings between them. The model is restricted to bulk homogeneous copolymers, or those copolymers dissolved in appropriate solvents without microphase separation. ↗

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## Introduction

Block and graft copolymers exhibit interesting viscoelastic properties. Although most of these systems show microphase separation, some of them are homogeneous, even in bulk, e.g., the block copolymers of styrene and  $\alpha$ -methyl styrene (1,2). Block copolymers in appropriate solvents also show no microphase separation (3-5). The molecular dynamics of these homogeneous copolymers may be treated on the basis of the bead-and-spring model enunciated by Rouse (6), Bueche (7), and Zimm (8). This theory has previously been applied to block copolymers of different configurations and compositions (9-15). In this paper, we extend the model to calculate the maximum relaxation times of graft copolymers and radial block copolymers. Viscoelastic relaxation times are computed numerically for these copolymers with different configurations.

## Theory

Following the method of analysis used in the calculations of block copolymers (11), we adopt a simplified procedure first proposed by Peticolas (16). The free-draining case of Rouse (6) and Bueche (7) is also used in our calculations.

The RBZ model represents the polymer molecule by  $N+1$  beads held together with  $N$  entropy springs. In a velocity gradient, the springs are stretched. The spring constant is  $3kT/b^2$  where  $b^2$  is the mean square end-to-end distance of the submolecule. As the beads move through the medium, a viscous drag is exerted on them the magnitude of which is determined by the friction co-

efficient  $f$ . At the cessation of flow, the recovery of the polymer chain proceeds without acceleration, so that the viscous and the elastic forces are exactly equal to each other. The equation of motion for the string of identical beads is given in matrix notation as (1)

$$\dot{\underline{x}} = -\sigma \underline{Z} \underline{x}, \quad (1)$$

where  $\underline{x}$  and  $\dot{\underline{x}}$  are the column vectors of the bead position and the bead velocity respectively,  $\underline{Z}$  is the nearest neighbor matrix and  $\sigma = 3kT/\langle b_0^2 \rangle f$  in which  $\langle b_0^2 \rangle$  is the mean square end-to-end distance and  $f$  is the frictional coefficient.

Eq. 1 was written for homopolymers. For graft copolymers, the equation must be modified to take into account the fact that not all beads are the same. Both the submolecular spring constants and the frictional coefficients of the beads are different (9). In addition, any bead connected to a graft point experiences elastic spring forces from all other beads immediately attached to the graft point and one other bead that is connected to this bead. To illustrate, bead  $k$  when disturbed from its equilibrium position (Figure 1) is pulled by three other beads,  $i, j, l$ . This gives the value 3 for the diagonal element  $(k, k)$  and -1 at off-diagonal positions  $(i, k)$ ,  $(k, i)$ ,  $(j, k)$ ,  $(k, j)$ ,  $(k, l)$ ,  $(l, k)$ , of the nearest neighbor matrix  $\underline{Z}$ . The  $\underline{Z}_g$  given below has 4's for some of its diagonal elements, because the matrix is written for graft copolymers with only one bead separating adjacent grafts. These beads are connected to four other beads, two on the grafts, the other two along the main chain. Thus, the resulting modified nearest neighbor matrix  $\underline{Z}_g$  can be written as:

[illegible]



For a graft copolymer consisting of side chains of polymer A, and a main chain of polymer B with frictional coefficients  $f_A$ ,  $f_B$ , respectively, the equations of motion become:

$$\dot{\underline{x}} = -\sigma_B \underline{D}^{-1} \underline{z}_g \underline{x}, \quad (2)$$

where  $\sigma_B = 3kT/b^2 f_B$ .  $\underline{D}^{-1}$  is the inverse of a diagonal matrix  $\underline{D}$  whose elements are the frictional coefficients of the corresponding beads divided by those of B beads. For example,

$$\underline{D} = \begin{bmatrix} 1 & & & & \\ & \ddots & & & \\ & & \delta_A & & \\ & & & \delta_A & \\ & & & & 1 & \\ & & & & & \ddots \\ & & & & & & \delta_A \\ & & & & & & & \delta_A \\ & & & & & & & & \ddots \end{bmatrix} \quad (3)$$

where  $\delta_A = b_A^2 f_A / b_B^2 f_B$ , occurring at positions corresponding to the side chain beads. The  $\underline{D}^{-1} \underline{z}_g$  matrix is no longer a tri-diagonal matrix.

To solve eq. 3., we define a new set of coordinates  $\underline{r}$  related to  $\underline{x}$  as follows:

$$\underline{r} = \underline{A} \underline{x}, \quad (4)$$

where the  $\underline{A}$  matrix is defined by  $\underline{z}_g = \underline{A}^T \underline{A}$ . Operating on both sides of eq. 4 with  $\underline{A}$ , it becomes

$$\begin{aligned} \dot{\underline{r}} &= -\sigma_A \underline{A} \underline{D}^{-1} \underline{A}^T \underline{A} \underline{x} \\ &= -\sigma_A \underline{S} \underline{r}, \end{aligned} \quad (5)$$

where

$$\underline{S} = \underline{A} \underline{D}^{-1} \underline{A}^T \quad (6)$$

is a symmetrical tridiagonal  $N \times N$  matrix which differs from the well known Rouse matrix ( $\underline{R} = \underline{A} \underline{A}^T$ ) by the extra operation  $\underline{D}^{-1}$  (6). For a homopolymer,  $\underline{D}$  matrix (eq. 3) is a unit matrix so that the  $\underline{S}$  matrix is reduced to the  $\underline{R}$  matrix. The Rouse model predicts for the latter case a discrete retardations spectrum (11):

$$\tau_p = 1/\sigma \lambda_p = f b^2 / 3 k T \lambda_p, \quad (7)$$

where  $\lambda'_p$ 's are the  $N$  eigenvalues of the  $\underline{R}$  matrix (6):

$$\lambda_p = 4 \sin^2 [p\pi/2(N+1)] , \quad p = 1, 2, \dots, N. \quad (8)$$

For graft copolymers, the eigenvalue spectrum of the  $\underline{S}$  matrix is readily obtained numerically on a computer by using a subroutine for determining the eigenvalues and eigenvectors of symmetric matrices. Retardation times are then calculated from these eigenvalues using eq. 7. The relaxation times have been shown to be just half of the corresponding retardation times (16).

The computed data are given in terms of normalized relaxation times defined by

$$\tau_p(\text{normalized}) = 1/(N+1)^2 \lambda_p \quad (9)$$

The use of the normalized relaxation time removes the arbitrariness of the choice of the value of  $N$  in the computation of maximum relaxation times. To arrive at eq. 9, we note that  $f = m\xi$ , where  $m$  is the number of monomers per submolecule and  $\xi$  is the monomeric friction coefficient, and



that  $b^2 \propto m\ell^2$  where  $\ell$  is the length of the monomeric segment. For a given molecular weight  $M$ ,  $m = M/(N + 1)$ . Therefore the relaxation time (eq. 7) must be inversely proportional to  $(N + 1)^2$ . The use of  $\tau_p$  (normalized), however, does not eliminate the dependence on  $N$  of shorter relaxation times (see eq. 8 for the case of homopolymers). Maximum relaxation time for a graft copolymer is normalized by that for a homopolymer as follows:

$$\tau_{\max}(\text{normalized}) = \tau_{\max \text{ graft}} / \tau_{\max \text{ homopolymer}} \quad (10)$$

For a radial block copolymer, similar equations can be set up to solve for the relaxation times. A schematic diagram for such a polymer is shown in Figure 2, which has  $r$  branches, each of which is consisted of  $g$  A-beads and  $m$  B-beads. The important distinction here is that beads connected to the radiating center are pulled by many other beads also attached to the radiating center. This gives rise to a value  $r$ , the number of radiating chains, at diagonal positions of the matrix  $\underline{Z}_r$  corresponding to these beads, and the value  $-1$  for all off-diagonal elements representing the interactions between any connected bead-pairs:



### Results and Discussion

To facilitate presentation and discussion of the results, the notation used in this work needs to be clarified. A general graft copolymer structure is depicted in Fig. 3, which shows a five-combed graft copolymer where  $g$  is the number of beads in the side chain, and  $m$  is the number of beads between two adjacent graft points.

The effects of varying the side chain lengths,  $g$ , and the spacings between the side chains,  $m$ , on the relaxation times have been investigated. Furthermore, the effect of having side chains of varying lengths on a given parent polymer and irregular spacings between adjacent graft points, in contrast to cases with uniform side chain length and regular spacing, has also been examined. In all instances,  $\delta_A$  was chosen to be 423. This value is obtained from literature data for A-beads of poly( $\alpha$ -methylstyrene) and B-beads of Polystyrene (1). The side chains were assumed to be poly( $\alpha$ -methylstyrene) and the parent polymer polystyrene.

The normalized relaxation times are plotted against the weight percent of polymer B (the backbone polymer, polystyrene) in Figure 4. In computing the weight percent, the value  $m_A/m_S=1.4$  is assumed (1), where  $m_A, m_S$  are the number of monomer units per submolecule of poly( $\alpha$ -methylstyrene) and polystyrene, respectively. The effects of introducing grafted chains with a higher friction coefficient are similar to those in the case of block copolymers (11). There is a split in the relaxation spectra, with the maximum relaxation times shifted to higher values.



The computed maximum relaxation times of the graft copolymers were normalized by those of the pure comb-shaped (17) homopolymer, polystyrene, as follows (1):

$$\tau_{\max}(\text{normalized}) = \frac{1}{2\sigma_B \lambda_{\min}} / \frac{1}{2\sigma_B \lambda_{\min}^{\circ}} = \frac{\lambda_{\min}^{\circ}}{\lambda_{\min}} \quad (11)$$

where  $\lambda_{\min}$  is the smallest of the eigenvalues, and  $\lambda_{\min}^{\circ}$  refers to that of the homopolymer. Table 1 presents calculated data on comb-shaped graft copolymers with different number of side chains (grafts). Fig. 5 exhibits a smooth curve drawn through 35 calculated points of different side chain lengths and graft spacings (and thus different weight compositions). These calculated cases also cover graft copolymers with three to ten side chains. All points follow the representative curve closely with a maximum scatter of less than 0.08 in  $\log \tau_{\max}$ . The difference vanishes as the number of side chains increases. This curve is therefore expected to give a good prediction of the maximum relaxation time behavior for polystyrene grafted with poly( $\alpha$ -methylstyrene) for any given number of side chains.

To further investigate the validity and generality of this computed curve, several other cases were calculated where the side chains were assumed to have different number of beads with the total number of side chain beads remaining constant, i.e.,  $g$  was not a constant but the sum of all the  $g$ 's was a constant (weight composition did not change, just the side chain lengths were irregular). Similarly, the number of beads separating the graft points was varied along the main chain, i.e.,  $m$  was not a constant while the sum of  $m$ 's was a constant

Finally, both  $g$  and  $m$  were set as variables. The results are tabulated in Table 2. The maximum relaxation time is seen to be an insensitive function of the distribution of side chain lengths and spacings between graft points. This finding further substantiates our previous conclusion that the maximum relaxation time as a function of graft copolymer composition can be reasonably represented by Fig. 5.

The viscoelastic behavior of radial multichain block copolymer can also be similarly computed. As shown in Fig. 2,  $g$  and  $m$  are numbers of beads in the poly( $\alpha$ -methylstyrene) and polystyrene blocks respectively. Calculations have been conducted for the case in which  $g$  and  $m$  are both 4, and the number of chains is 9, as a preliminary attempt to determine if multichain radial block copolymers behave differently from comb-shaped graft copolymers. The results for a polymer containing 41.7% polystyrene are given in Table 3. The normalized maximum relaxation time is  $3.921 - 1.360 = 2.561$  in log scale. This result suggests that the behavior of a multichain block copolymer resembles that of ASA triblock copolymer (1,11) where A and S stand for poly( $\alpha$ -methylstyrene) and polystyrene blocks. Here, varying the distributions of  $g$ 's and  $m$ 's is seen to result in significant changes in the maximum relaxation times regardless of the fact that the weight composition has been preserved.

Table 4 shows the calculated viscoelastic parameters for 15 multichain radial block copolymers. These block copolymers are star-shaped with branches of equal length. The normalized relaxation times are plotted against the weight percent of styrene



in Figure 6. From the calculated maximum relaxation times of these cases and those of the corresponding branched homopolymers (17), the normalized maximum relaxation time can be plotted as a function of the weight percent of styrene blocks as shown in Fig. 7. A smooth curve can be drawn through all calculated points. This curve shows the same general trend as Fig. 5 but the rise is much sharper as the  $\alpha$ -methylstyrene weight content begins to increase. It appears that the end  $\alpha$ -methylstyrene blocks are very effective in increasing the maximum relaxation time of the multichain radial block copolymer.

In conclusion, the maximum relaxation times as a function of composition for graft and radial block copolymers have been computed (Figs. 5 and 7). These curves show the relative retardation effectiveness of the higher friction beads placed at different locations in the macromolecules. These beads when grafted onto the side of a linear main chain (lower friction beads) are less effective than those added to the end of the branches of a star-shaped parent polymer. Note that Fig. 7 resembles the ABA curve by Hansen and Shen (1,2) which is to be expected since ABA is the limiting case of the radial block copolymers.

#### Acknowledgement

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Table 1. Viscoelastic Parameters of Comb-shaped Graft Copolymers

Number of Grafts	g	m	Log $\tau_{\max}$	$\eta_0$	$J_{eR}$
5	1	5	3.734	$8.68 \times 10^3$	0.442
5	1	7	3.864	$1.16 \times 10^4$	0.449
5	1	9	3.982	$1.51 \times 10^4$	0.453
5	1	11	4.075	$1.86 \times 10^4$	0.456
5	2	5	4.016	$1.81 \times 10^4$	0.381
5	3	5	4.205	$2.99 \times 10^4$	0.340
5	4	5	4.343	$4.37 \times 10^4$	0.310
5	5	5	4.452	$5.93 \times 10^4$	0.287
5	5	4	4.360	$5.08 \times 10^4$	0.264
5	5	3	4.244	$4.25 \times 10^4$	0.236
5	5	2	4.087	$3.42 \times 10^4$	0.202
5	5	1	3.851	$2.59 \times 10^4$	0.165
6	1	4	3.751	$9.36 \times 10^3$	0.413
6	2	4	4.063	$2.10 \times 10^4$	0.350
6	3	4	4.251	$3.49 \times 10^4$	0.308
6	4	4	4.388	$5.11 \times 10^4$	0.277
6	4	3	4.261	$4.15 \times 10^4$	0.247
6	4	2	4.085	$3.19 \times 10^4$	0.205
6	4	1	3.791	$2.21 \times 10^4$	0.151
3	1	5	3.297	$2.74 \times 10^3$	0.595
3	2	5	3.623	$6.48 \times 10^3$	0.492
3	3	5	3.829	$1.12 \times 10^4$	0.437
3	4	5	3.983	$1.69 \times 10^4$	0.401
3	5	5	4.107	$2.37 \times 10^4$	0.376
3	5	4	4.033	$2.08 \times 10^4$	0.355
3	5	3	3.943	$1.80 \times 10^4$	0.330
3	5	2	3.834	$1.53 \times 10^4$	0.303
3	5	1	3.699	$9.23 \times 10^3$	0.275
10	1	3	4.049	$1.92 \times 10^4$	0.378
10	2	3	4.357	$4.25 \times 10^4$	0.322
10	3	3	4.540	$6.91 \times 10^4$	0.281
10	3	2	4.342	$4.93 \times 10^4$	0.235
10	3	1	3.974	$2.86 \times 10^4$	0.148



Table 2. Viscoelastic Parameters of Graft Copolymers  
with a Distribution of Side Chain Lengths, g,  
and Spacing Between Graft Points, m.

g	m	Log $\tau_{\max}$	$\eta_0$	$J_{eR}$
5,5,5,5,5	all 5's	4.452	$5.93 \times 10^4$	0.287
4,3,5,11,2	all 5's	4.461	$5.78 \times 10^4$	0.301
4,1,10,9,1	all 5's	4.390	$5.73 \times 10^4$	0.287
5,4,7,2,7	all 5's	4.494	$6.25 \times 10^4$	0.315
4,7,6,6,2	all 5's	4.381	$5.53 \times 10^4$	0.258
all 5's	11,8,1,1,4	4.525	$6.00 \times 10^4$	0.356
all 5's	17,4,1,1,2	4.550	$6.00 \times 10^4$	0.383
all 5's	7,9,1,1,7	4.492	$5.60 \times 10^4$	0.349
13,1,7,1,3	6,3,5,10,1	4.596	$7.03 \times 10^4$	0.367
4,4,5,7,5	1,6,5,2,11	4.393	$5.00 \times 10^4$	0.298
10,9,1,1,4	3,12,4,5,1	4.575	$7.10 \times 10^4$	0.356

Table 3. Effects of Distribution of Block Lengths,  $g$ ,  
 $m$ , on the Viscoelastic Parameters of Radial  
 Block Copolymers (Number of chains = 9)

$g$	$m$	$\text{Log } \tau_{\text{max}}$	$\eta_0$	$J_{eR}$
all 4's	all 4's	3.921	$7.52 \times 10^4$	0.0993
5,10,1	all 4's	4.395	$8.24 \times 10^4$	0.155
1,3,6				
3,5,2				
all 4's	13,9,1			
	1,1,1	4.346	$7.73 \times 10^4$	0.161
	1,7,2			
1,1,7	5,1,3			
5,6,4	18,2,3	4.564	$8.74 \times 10^4$	0.225
2,7,3	1,1,2			



Table 4. Viscoelastic Parameters of Multichain Radial  
Block Copolymers

Number of Branches	g	m	Log $\tau_{\max}$	$\eta_0$	$J_{eR}$
5	1	5	3.344	$8.86 \times 10^3$	0.248
5	1	7	3.486	$1.23 \times 10^4$	0.247
5	1	9	3.593	$1.58 \times 10^4$	0.247
5	1	11	3.679	$1.92 \times 10^4$	0.246
5	2	5	3.665	$1.96 \times 10^4$	0.224
5	3	5	3.866	$3.21 \times 10^4$	0.211
5	4	5	4.016	$4.65 \times 10^4$	0.201
5	5	5	4.138	$6.27 \times 10^4$	0.193
5	5	4	4.066	$5.42 \times 10^4$	0.187
5	5	3	3.982	$4.57 \times 10^4$	0.178
5	5	2	3.878	$3.72 \times 10^4$	0.168
5	5	1	3.748	$2.88 \times 10^4$	0.156
3	1	5	3.355	$4.55 \times 10^3$	0.495
3	2	5	3.676	$1.01 \times 10^4$	0.440
3	3	5	3.876	$1.67 \times 10^4$	0.408
3	4	5	4.026	$2.42 \times 10^4$	0.386
3	5	5	4.146	$3.28 \times 10^4$	0.369
3	5	4	4.076	$2.85 \times 10^4$	0.354
3	5	3	3.994	$2.43 \times 10^4$	0.335
3	5	2	3.893	$2.00 \times 10^4$	0.312
3	5	1	3.767	$1.58 \times 10^4$	0.284
10	1	3	3.118	$1.18 \times 10^4$	0.111
10	2	3	3.455	$2.77 \times 10^4$	0.0962
10	3	3	3.673	$4.74 \times 10^4$	0.0893
10	3	2	3.539	$3.60 \times 10^4$	0.0844
10	3	1	3.353	$2.46 \times 10^4$	0.0775
3	1	15	3.817	$1.33 \times 10^4$	0.486

Figure Captions

- Figure 1. Modelling of a graft point. Bead  $k$  experiences forces from beads  $i$ ,  $j$  and  $l$ , and vice versa.
- Figure 2. Schematic diagram of a radial multichain block copolymer where  $g$  is the number of poly( $\alpha$ -methylstyrene) beads, and  $m$  is the number of polystyrene beads in the respective blocks.
- Figure 3. Schematic diagram of a graft copolymer with 5 grafts along the parent chain.  $g$  is the number of beads in the side chains.  $m$  is the number of beads of the section separating two adjacent graft points.
- Figure 4. Normalized relaxation time spectra for a series of comb-shaped graft copolymers with varying PS contents.
- Figure 5. Normalized maximum relaxation time as a function of weight percentage of styrene for polymers of styrene grafted with poly( $\alpha$ -methylstyrene). Circles are calculated points.
- Figure 6. Normalized relaxation time spectra for a series of radial block copolymers with varying PS contents.
- Figure 7. Normalized maximum relaxation time as a function of weight percentage of styrene for multichain radial block copolymers with  $\alpha$ -methylstyrene end blocks. Circles are calculated points.

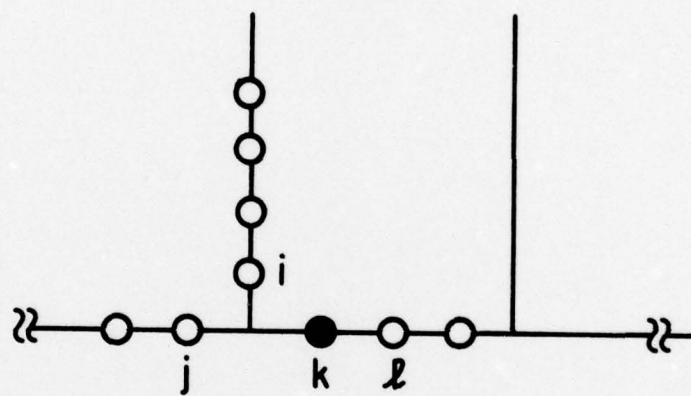


Figure 1

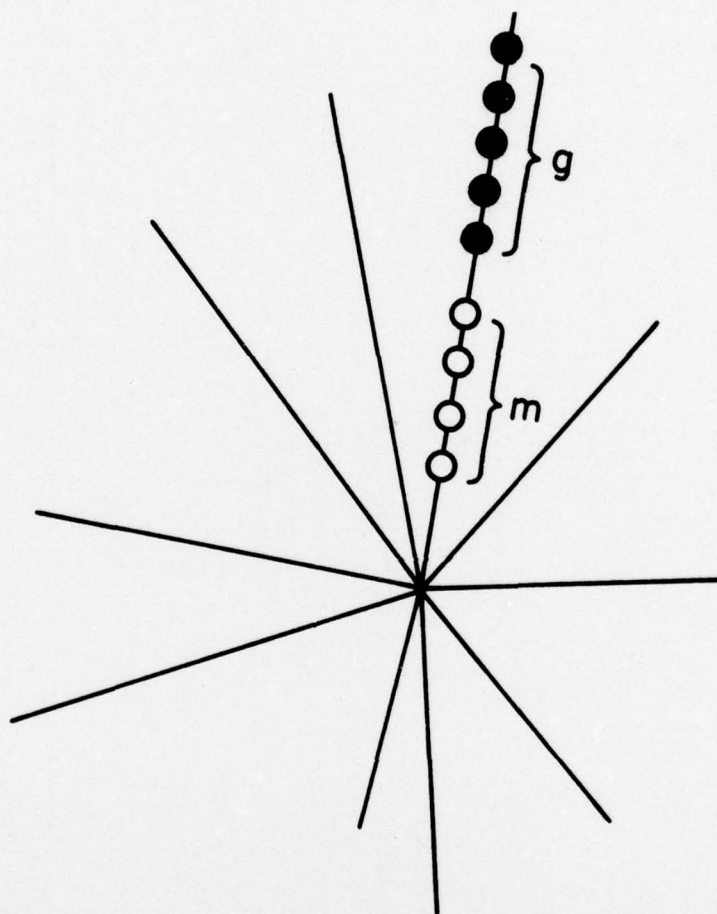


Figure 2



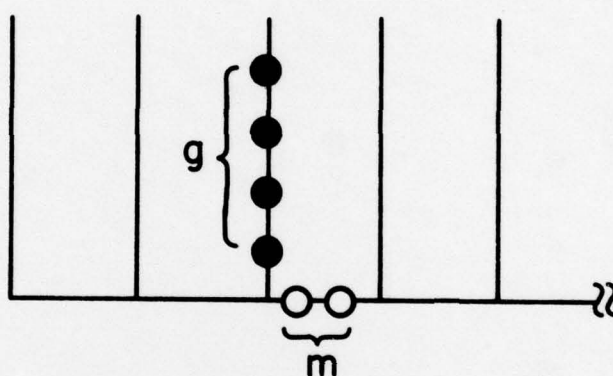


Figure 3



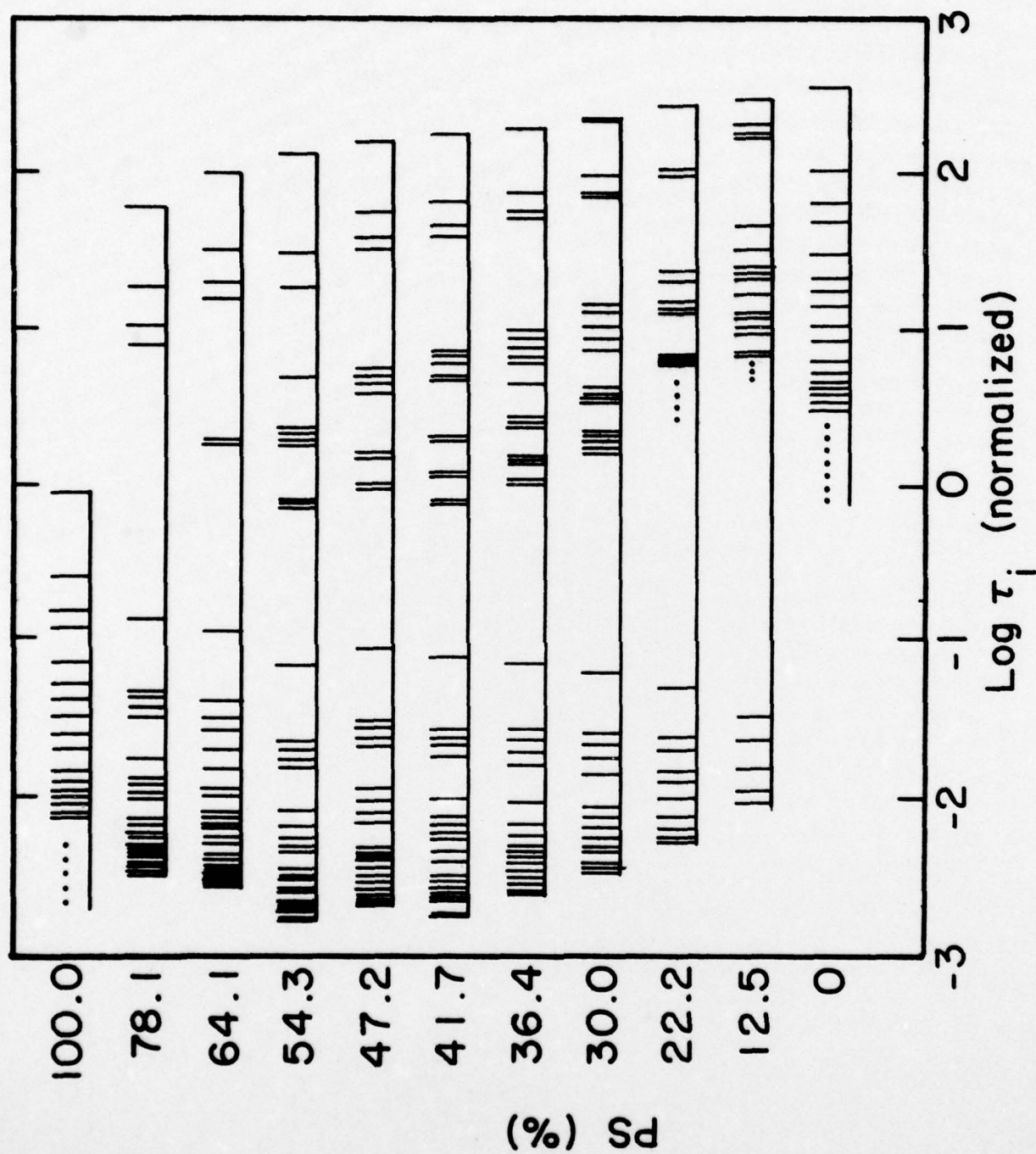


Figure 4

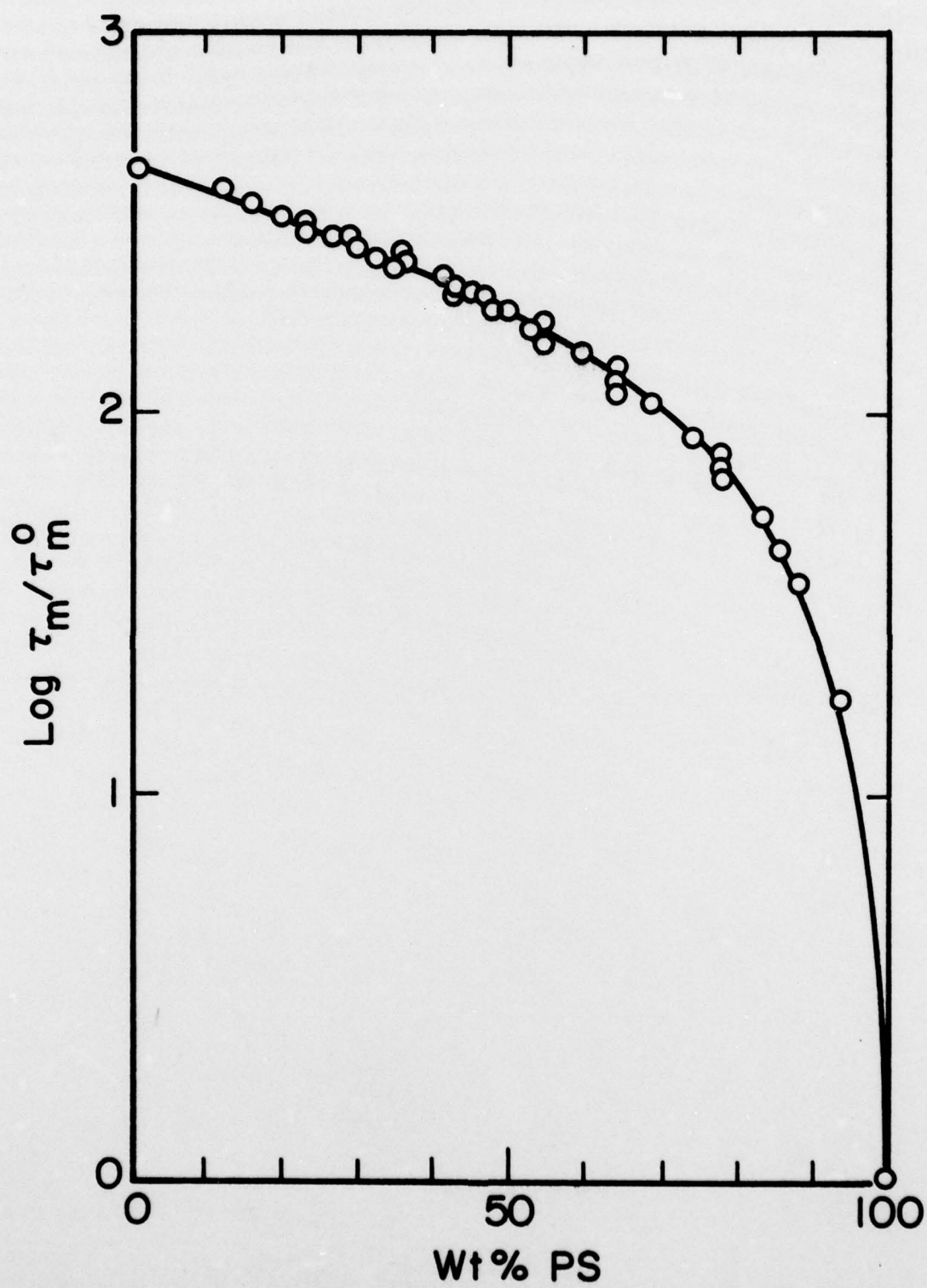


Figure 5

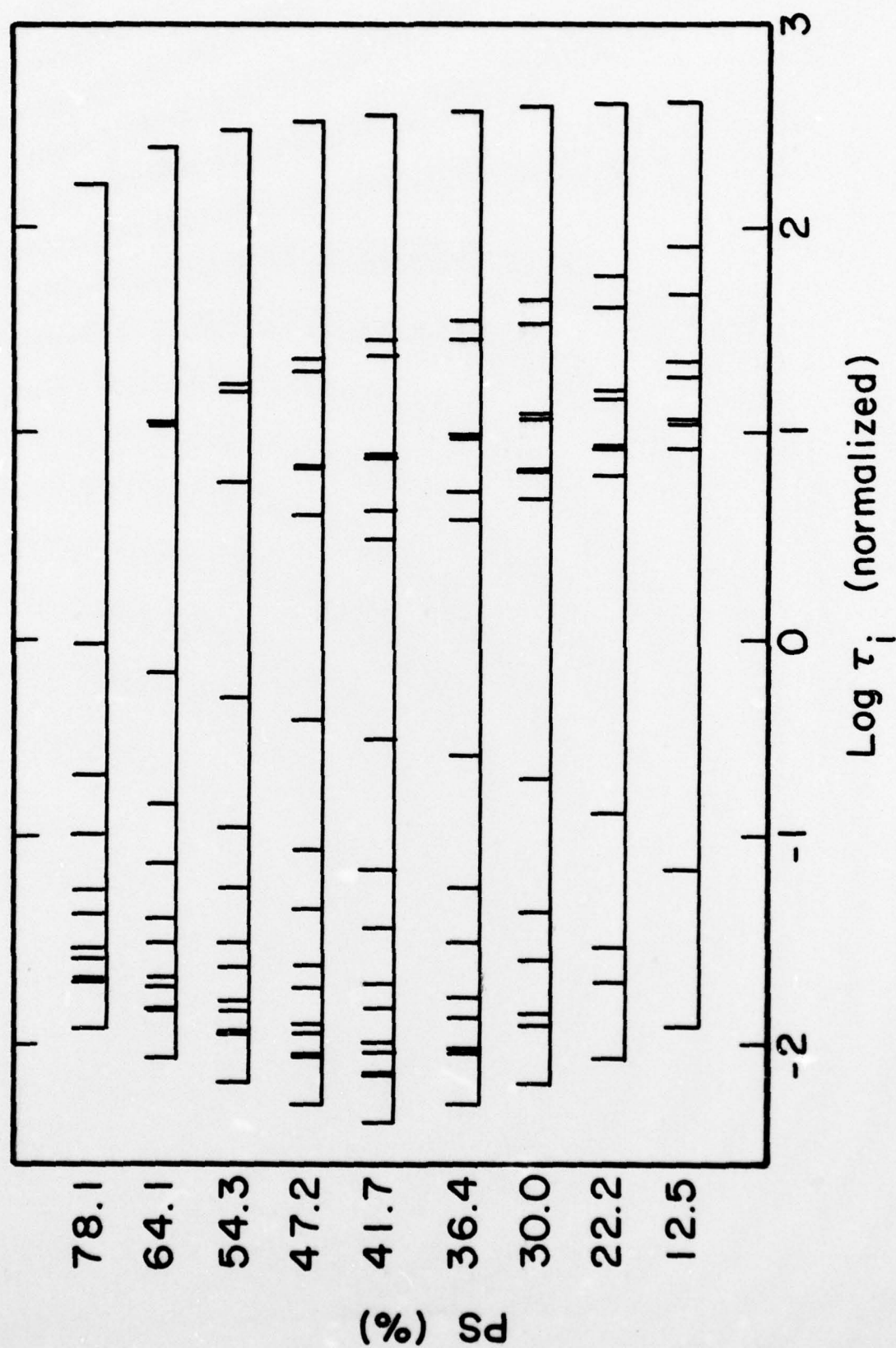


Figure 6



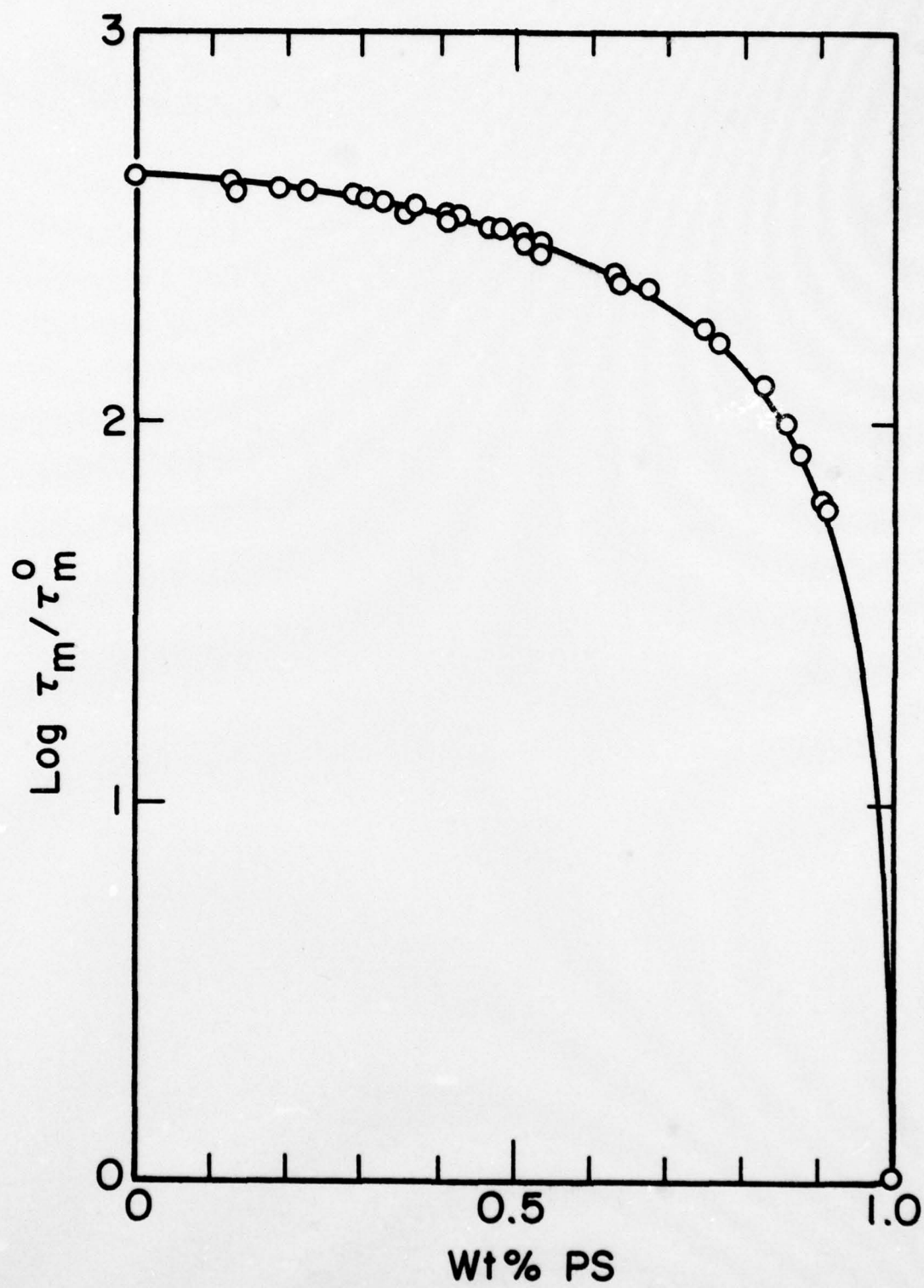


Figure 7

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